

09/7625 13

Atty. Docket #: 1998/F-751

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

INTERNATIONAL APPL. NO.: PCT/EP99/04570 :

INTERNATIONAL FILING DATE: -07/01/1999- :

APPLICANT: ULRICH GEBHARDT ET AL :

SERIAL NO: : **ART UNIT:**

FILED: -HEREWITH- : **EXAMINER:**

FOR: "PEM FUEL CELL WITH IMPROVED
LONG-TERM PERFORMANCE, METHOD FOR
OPERATING A PEM FUEL CELL AND PEM
FUEL CELL STORAGE BATTERY" :

Commissioner for Patents

Box PCT

Washington, D.C. 20231

"Express Mail" No.: EE617838639

Date: -FEBRUARY 08, 2001-

I hereby certify that this paper, along with any other paper or fee referred to in this paper as being transmitted herewith, is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10, postage prepaid, on the date indicated above, addressed to the Commissioner for Patents, Washington, D.C. 20231

- Amy L. Hamm -

(Typed or printed name of mailing paper or fee)

Amy L. Hamm
(Signature of person mailing paper)

**TRANSMITTAL OF APPLICATION PAPERS
TO U.S. DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. §371
(37 CFR 1.494 OR 1.495)**

This Transmittal Letter is based upon PTO Form 1390 (as revised in May, 1993).

The above-identified applicant(s) (jointly with their assignee) have filed an International Application under the P.C.T. and hereby submit(s) to the United States Designated/Elected Office (DO/EO/US) the following items and other information.

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. §371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. §371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. §371(f)) at any time rather than delay.
4. ☒ A proper Demand for International Preliminary Examination (IPE) was made to the appropriate Authority (IPEA) within the time period required.
5. ☒ A copy of the International Application as filed (35 U.S.C. §371(c)(2)) --
 - a. ☒ is transmitted herewith (required when not transmitted by International Bureau).
 - b. ☐ has been transmitted by the International Bureau. See WIPO Publication WO 00/10215.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A (verified) translation of the International Application into the English language is enclosed -with- One (1) Sheet of Drawings.
7. ☐ Amendments to the (specification and) claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are transmitted herewith (required if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☐ have not been made and will not be made.
 - e. ☐ will be submitted with the appropriate surcharge.
8. ☐ A translation of the amendments to the claims (and/or the specification) under PCT Article 19 (35 U.S.C. §371(c)(3)) is enclosed or will be submitted with the appropriate surcharge.

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International Application No. PCT/EP99/04570

9. ☒ An oath or declaration/power of attorney of the inventor(s) (35 U.S.C. §371[c][4]) will follow.
☐ and is attached to the translation of (or a copy of) the International Application.
☐ and is attached to the substitute specification.

10. ☐ A translation of at least the Annexes to the IPE Report under PCT Article 36 (35 U.S.C. §371[c][5]) is enclosed.

Items 11. to 16. below concern other document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98 is enclosed.
12. ☒ An Assignment for recording and a separate cover sheet in compliance with 37 CFR 3.28 and 3.31 will follow.
13. ☒ A FIRST preliminary amendment is enclosed.
A SECOND or SUBSEQUENT preliminary amendment is enclosed.
14. ☐ A substitute specification (including claims, abstract, drawing) is enclosed.
15. ☐ A change of power of attorney and/or address letter is enclosed.
16. ☒ Other items of information:

- ☒ This application is being filed pursuant to 37 CFR 1.494(c) or 1.495(c), and any missing parts will be filed before expiration of--

☐ 22 months from the priority date under 37 CFR 1.494(c), or

☒ 32 months from the priority date under 37 CFR 1.495(c).

- ☒ The undersigned attorney is authorized by the International applicant and by the inventors to enter the National Phase pursuant to 37 CFR 1.494(c) or 1.495(c).

The following additional information relates to the International Application:

International Application No. PCT/EP99/04570

1998/F-751

- ☒ Receiving Office: EPO
☒ IPEA (if filing under 37 CFR 1.495): EPO
☒ Priority Claim(s) (35 USC §§ 119, 365):
198 36 142.4 filed August 10, 1998 -and-
German Appln. 298 15 330.0 filed August 26, 1998.
☒ A copy of the International Search Report is

☐ enclosed.

☒ attached to the copy of the International
Application.

☒ A copy of the Receiving Office Request Form is enclosed. (In German)

☒ Letter of March 06, 2000 to WIPO (1) sheet.

The fee calculation is set forth on the next page of this Transmittal Letter.

FEE CALCULATION SHEET

☒ A check in payment of the filing fee, calculated as follows, is attached (37 CFR 1.492).

Basic Fee..... \$ 860.00

Total Number of claims in
excess of (20) times \$18..... -0-

Number of independent claims
in excess of (3) times \$80..... -0-

Fee for multiple dependent
claims \$270..... -0-

TOTAL FILING FEE... \$ 860.00

Kindly send us the official filing receipt.

The Commissioner is hereby authorized to charge any additional fees which may be required or to credit any overpayment to Deposit Account No. 03-2775. This is a "general authorization" under 37 CFR 1.25(b), except that no automatic debit of the issue upon allowance is authorized. An additional copy of this page is attached.

Respectfully submitted,

By Richard M. Beck

Richard M. Beck

Reg. No. 22,580

CONNOLLY BOVE LODGE & HUTZ LLP

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RMB/alh

Enclosures

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(8577*25)

1998/F-751

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

ULRICH GEBHARDT ET AL : ART UNIT:
SERIAL NO.: :
FILED: -HEREWITH- : EXAMINER:
TITLE: "PEM FUEL CELL WITH IM- :
PROVED LONG-TERM PERFORMANCE, :
METHOD FOR OPERATING A PEM FUEL :
CELL AND PEM FUEL CELL STORAGE :
BATTERY" :

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Date of Deposit -Feb. 08, 2001-
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Service "Express Mail Post Office to
Addressee" service under 37 CFR 1.10 on the
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PCT, Commissioner for Patents,
Washington, D.C. 20231.
- Amy L. Hamm -
(Typed or printed name of person mailing
paper or fee)
Amy L. Hamm
(Signature of person mailing paper or fee)

Commissioner for
Patents
Washington, D.C. 20231

PRELIMINARY AMENDMENT

Sir:

Prior to the examination of this application, the applicant(s)
respectfully request(s) that this Preliminary Amendment be entered.

In the Claims:

Claim 4, line 4, change "one of claims 1 to 3" to read --
claim 1 -- .

R E M A R K S

Claim 4 has been amended to refer to only one preceding
claim. Each of the dependent claims, as amended, now depends
on only one preceding claim. Therefore no additional fee is
required for multiple dependency.

Prompt, favorable action is solicited.

Respectfully submitted,

CONNOLLY BOVE LODGE & HUTZ LLP

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RMB/des
(8577*25)

Description

5 PEM FUEL CELL WITH IMPROVED LONG-TERM PERFORMANCE,
METHOD FOR OPERATING A PEM FUEL CELL AND PEM FUEL CELL
STORAGE BATTERY

10 The invention relates to a polymer electrolyte membrane (PEM) fuel cell with a new type of design of the edge region, and to a method for operating a fuel cell and to a fuel cell storage battery.

15 A design of an edge region of a PEM fuel cell in which the edge seal is made by a frame element which presses the respectively adjacent collector plate onto the top and bottom of the membrane in such a way that the three parts are connected to one another in a mechanically secure, gastight and electronically insulating manner, is known from DE-C 44 42 285 (see Fig. 2 therein). The two electrodes with which the membrane is coated on
20 each side do not extend as far as into this edge region. Therefore, there is a minute gap formed at the boundary between the electrode coating of the membrane and the edge seal, at which gap the membrane is directly exposed to the process gases, i.e. without a
25 protective electrode layer. This causes the membrane to dry out and become brittle here. Also, previous damage which may be caused, for example, during the hot pressing of the membrane-electrodes unit may lead to gas breakthroughs at this location where the membrane is directly exposed to the process gases. The
30 utilization time or long-term performance of a membrane is correspondingly limited by this gap at which the membrane is directly exposed to the process gases.

35 It is an object of the present invention to provide a PEM fuel cell structure with improved long-term performance.

This object is achieved by a PEM fuel cell as claimed in claim 1, by the method for its production as claimed in claim 3 and by the provision of the fuel cell storage battery as claimed in claim 4. Further
5 configurations of the invention are given in the description, the figures and the explanations thereof.

The invention relates to a PEM fuel cell which comprises at least two terminal plates which clamp in a
10 membrane which is covered on both sides, apart from the outermost edge, by an electrode layer, the covering of the membrane with at least one electrode layer projecting into the structural edge region of the fuel cell. This enlargement of at least one electrode layer
15 not only means that the membrane, at least on one side, is no longer directly exposed to the process gas, but also even leads to a small reservoir of water being formed at the boundary between electrode-coated and uncoated membrane in the edge region, which water
20 reservoir continuously wets the membrane.

The invention also relates to a method for operating a PEM fuel cell, in which the formation of product water in the structural edge region of the fuel cell is
25 utilized to wet the membrane.

Finally, the invention relates to a PEM fuel cell storage battery, comprising at least two PEM fuel cells as claimed in one of claims 1 to 3.

30

In this context, the term terminal plate is understood as meaning any type of separators and cooling and contact plate which enclose the gas space of a fuel cell on the side which lies opposite the membrane.

35

The "structural edge region" of the fuel cell is understood as meaning that region of the cell which

lies outside the active cell areas and in which, therefore, there is no regular supply and removal of process gases and reaction products.

- 5 The electrode layer is a gas-permeable layer and preferably comprises an active catalyst layer and a support, such as for example a carbon paper.

10 The membrane is preferably a proton-conducting electrolyte film which in the operating state has a water content of approx. 20-40% by weight.

15 In the edge region, seals are preferably arranged between the terminal plates and the membrane.

20 According to one configuration of the invention, the frame element is made from metal and an electrically insulating layer is additionally present in the edge region, allowing series connection when the individual cells are stacked without there being any risk of a short circuit.

25 One configuration of the invention is explained below with reference to two figures, in which:

30 Figure 1 shows the structure of a fuel cell in cross section, and

Figure 2 shows a detailed enlargement of the edge region.

35 Figure 1 shows a fuel cell 1. The membrane 2, which extends over the entire length of the cell, is in the center. The membrane is coated on both sides with the electrodes 3 and 4, as far as the edge. The seals 5 and 6, which adjoin the two sides of the membrane where the electrodes stop, can be seen at the edge. The terminal plates 7 and 8, which delimit the two reaction spaces

11 and 12 of the fuel cell 1 on the opposite side from the membrane 2, can be seen at the top and bottom.

The cross section through the fuel cell 1 selected in Figure 1 is taken through the supply or removal ducts 9/10 for the process gases. Therefore, in each case two removal or supply openings, through which the process gases flow, for example in the direction indicated by the arrows, can be seen in the terminal plates 7 and 8.

10 The cell area between the supply and removal ducts is the active cell area. The edge region of the fuel cell lies on the other side of the ducts.

In operation, a process gas, for example the fuel, flows through the distribution ducts 13 into one of the two reaction spaces 11/12, for example the anode chamber 11, along the active cell area where the reaction of oxidant and fuel to form water and current takes place. The product water is regularly removed along the active cell area. Hitherto, the active cell area has been the only point in a fuel cell at which product water is formed. According to the invention, reaction now also takes place, to a slight extent, in the structural edge region of the cell, where the electrode layers have according to the invention been extended along the membrane. The process gases reach this area practically only by diffusion through the support of the active catalyst layer, i.e. for example through the carbon paper, since the terminal plates in the structural edge region do not have any distribution ducts 13.

As has been stated, the process gas flows in the structural edge region are small or even nonexistent and therefore the product water formed there cannot be removed. Consequently, product water 14 collects in the gap which forms and adjoins the end of the electrode layer on the membrane. As a result, a small reservoir

of water 14 is formed between the seals 5 and 6 and the membrane 2. This reservoir of water offers the following advantages:

- 5 1.) The membrane surface which lies outside the active electrode surface is always surrounded by water. Membranes whose mechanical resistance is highly dependent on the water content can therefore be used with long-term stability.
- 10 2.) Any damage which may be present in the edge region of the membrane, caused, for example, by hot pressing, could hitherto, i.e. without the reservoir of water, have led to gas breakthroughs. On account of the water
15 cushion which is now present, only gases which are dissolved in water can diffuse to the membrane. This quantity of gas is so small that there is no possibility of local overheating and further damage to the membrane, such as for example a gas breakthrough.
- 20 3.) The membrane is prevented from becoming brittle and drying out in the edge region.

The region which is circled in Figure 1 is shown in
25 detail in Figure 2. The membrane 2, which is surrounded by the seals 5 and 6 at the edge, is arranged in the center. Toward the center of the cell area, it is coated with the electrodes 3 and 4, which comprise the catalyst layers 3a and 4a and the supports 3b and 4b.
30 The axial supply duct 10, the terminal plates 7 and 8 with their distribution ducts 13 in the reaction spaces 11 and 12 can also be seen. A reservoir of water 14 is formed at the end of each of the electrode coatings of the membrane, since the product water which is formed
35 there cannot be removed.

The novel extension of the electrode layer into the structural edge region of the fuel cell means that a

reservoir of water, which wets the membrane, is formed in that region in a gap at a location on the membrane.

Patent claims

1. A PEM fuel cell which comprises at least two terminal plates which clamp in a membrane which, apart from the outermost edge, is covered on both sides by an electrode layer, the covering of the membrane with at least one electrode layer projecting into the structural edge region of the fuel cell.
2. The PEM fuel cell as claimed in claim 1, in which in the edge region seals are arranged between the membrane and the frame element.
3. A method for operating a PEM fuel cell, in which the formation of product water in the structural edge region of the fuel cell is utilized to wet the membrane.
4. A fuel cell storage battery which is made from fuel cells which are electrically series-connected and comprises a stack of at least two fuel cells as claimed in one of claims 1 to 3 which are mechanically connected to one another.

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[illegible][illegible][illegible]

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FIG 1

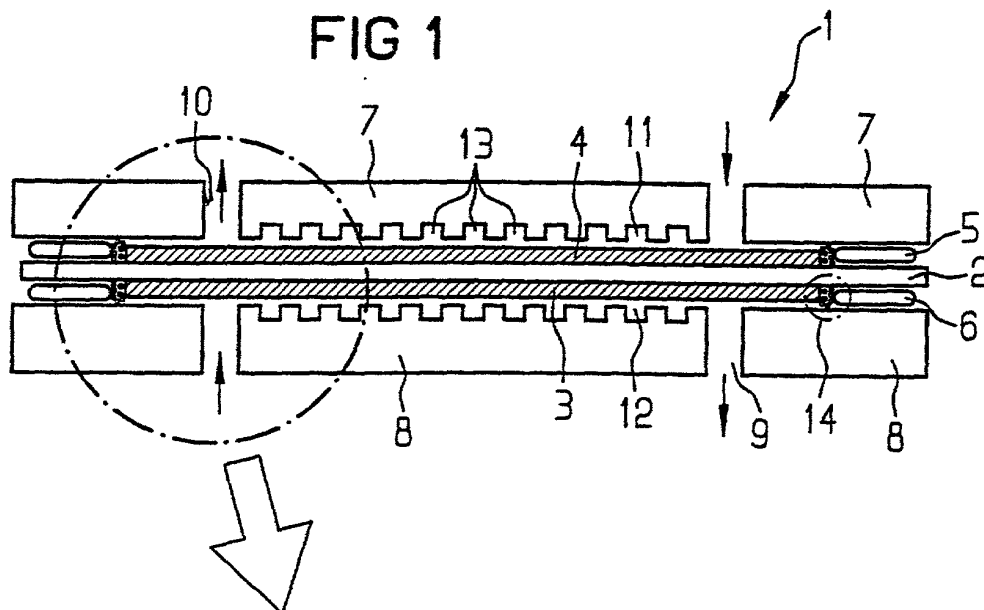
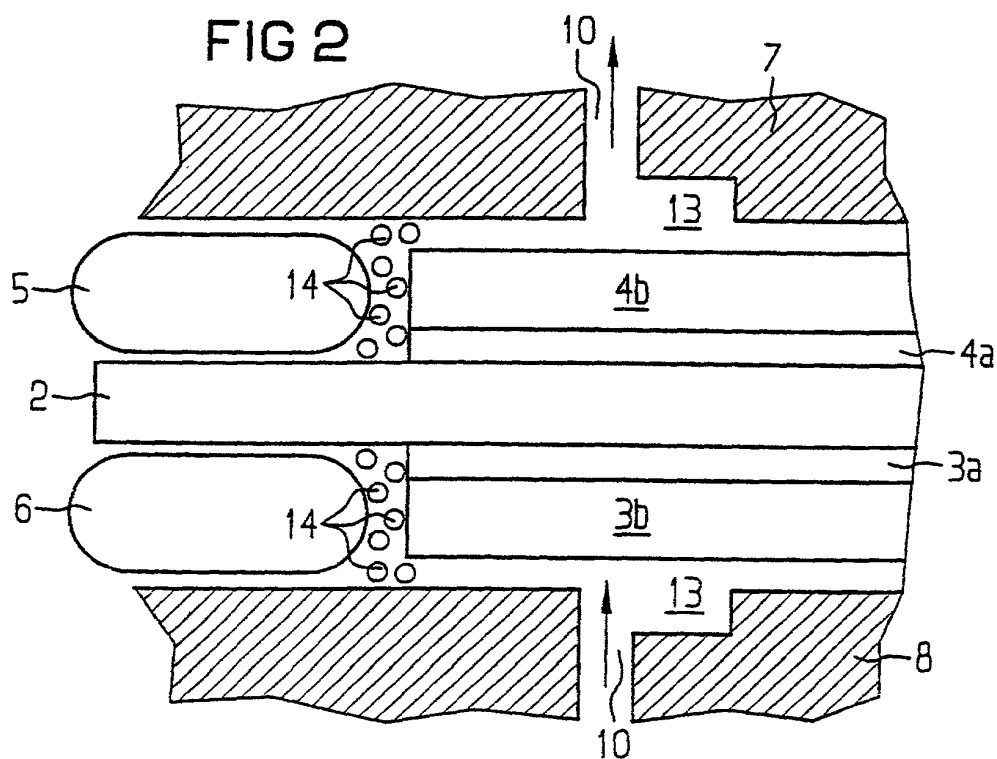


FIG 2





1998/F751 US

COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY

As below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below, I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

PEM fuel cell with improved long-term performance, method for operating a PEM fuel cell and PEM fuel cell storage battery

the specification of which

- is attached hereto

- was filed on July 7, 1999 as International Patent Application PCT/EP99/04570

and including all the amendments through the date hereof.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s) for which Priority is Claimed:

Federal Republic of Germany, 19836142.4 of August 10, 1998

Federal Republic of Germany, 29815330.0 of August 26, 1999

And I hereby appoint

Rudolf E. Hutz, Reg.No. 22,397; Harold Pezzner, Reg.No. 22,112; Richard M. Beck, Reg.No. 22,580; Paul E. Crawford, Reg.No. 24,397; Thomas M Meshbeshner, Reg.No. 25,083; Robert G. McMorrow, Jr., Reg.No. 30,962; Patricia Smink Rogowski, Reg.No. 33,791; Ashley I. Pezzner, Reg.No. 35,646; William E. McShane, Reg. 32,707; James T. Moore, Reg. No. 35,619; Mary W. Bourke, Reg.No. 30,982; Gerard M. O'Rourke, Reg.No. 39,794; Christine M. Hansen, Reg.No. 40,634; Allan N. Kutzenco, Reg. No. 38,945; James M. Olsen, Reg.No. 40,408; Francis DiGiovanni, Reg.No. 37,310; Frank Z. Yang, Reg. No. 35,417; Eric J. Vain, Reg.No. 42,517; Daniel C. Mulveny, Reg.No. P-45,897; and Elliot C. Mendelson, Reg.No. 42,878

all of CONNOLLY AND HUTZ, P.O.Box 2207, Wilmington, Delaware 19899-2007, my attorneys with full power of substitution, to prosecute this application, and transact all business in the Patent and Trademark Office connected therewith and I hereby request that all correspondence in this application be directed to:

CONNOLLY AND HUTZ

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Wilmington, Delaware 19899

Telephone (302) 658-9141

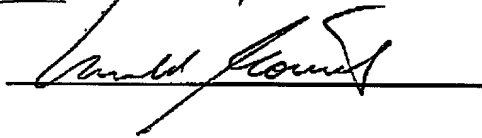
20

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

INVENTOR(S) / Residence

1) Dr. Harald Bönsel, Flachsweg 18, 65524 Niedernhausen, Germany

Signature:



Date:

02. Juli 2001

2) Dr. Gregor Deckers Johannesallee 41, 65929 Frankfurt, Germany

Signature:



Date:

27 JUN 2001

The inventors are citizens of Germany.

Post Office Address of all Inventors:

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